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**Process and apparatus for producing cling-fastener
parts made from radiation-cured plastics**

10 The present invention relates to a process and an apparatus for producing cling-fastener parts which have a large number of interlocking means.

15 Known cling-fastener parts are produced from thermoplastic polyolefins by extrusion. Here, the thermoplastic, in the plastic or liquid state, is fed, for example, to a gap between a pressure roll and a shaping roll, the shaping roll having a large number of radial cutouts, both ends of which are open. The 20 thermoplastic penetrates into the cutouts under the action of the nip pressure, and substantially cures, so that the cling-fastener parts, in the form of a three-dimensional structure, can be released from the shaping roll. The cling-fastener base is molded in the gap 25 between the shaping roll and the pressure roll, the base and the interlocking means, specifically the interlock projections formed in the cutouts and the interlock tips formed, being one single bonded piece.

30 The materials preferably used in the conventional processes are thermoplastics such as polypropylene, polyamide or polyethylene.

35 A process of this type is known from WO 98/20767, for example.

High nip pressures of from about 500 kilogram/m² to a few metric tons/m² are required in order to achieve adequate supply of the plastic material, in its plastic

or liquid state, to the cutouts.

In addition, the relatively low cooling rate of the thermoplastic polymers means that only small meterages 5 of the three-dimensional cling-fastener sheeting can be produced on a shaping roll of width about 400 mm.

Production of cling-fastener parts via extrusion of thermoplastics requires considerable energy cost, to 10 heat the thermoplastic composition to temperatures as high as 300°C.

The known processes place production-related 15 restrictions on both the maximum width of the web of cling-fastener sheet and the minimum thickness of the cling-fastener sheet, and the company Velcro Industrie B.V. has therefore developed longitudinal and/or transverse stretching processes, described in PCT WO 20 98/32349, for producing wider, and very thin, film-type cling-fastener sheet. A disadvantage with this process, besides the large amount of high-cost resource used for production, is that each stretching process markedly reduces the number of interlocking means per unit of area.

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The object of the present invention is to provide a novel process and a novel apparatus for producing cling-fastener parts, where these permit production rate to be increased while reducing energy cost. The 30 present invention is moreover intended to permit the provision of cling-fastener parts with greater heat resistance, and also of film structures with extremely low thickness, while providing a large number of protruding elements or interlocking means.

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Very surprisingly, it has been found possible to achieve a considerable rise in production rate, while reducing energy costs, by shaping, casting, and/or compression molding a formulation encompassing

radiation-crosslinkable, preferably acrylic, prepolymers, and then radiation-curing. It is also possible to dispense with the use of inert atmosphere if the radiation-crosslinkable prepolymers are suitably selected.

Exceptionally high polymerization rates are achieved in the radiation curing of formulations encompassing radiation-crosslinkable, in particular acrylic, prepolymers, where this takes place by way of UV radiation or electron beam. Compared with the known production processes for cling-fastener parts made from thermoplastics, the process of the invention can give a ten-fold increase in the rate of rotation of the rolls, and therefore in the length of the three-dimensional webs produced per unit of time.

Since polymerization by way of radiation crosslinking does not require any heating of the curable composition, as is required in the known processes, the process of the invention also saves energy.

Another advantage of radiation curing is that the polymerization takes place without releasing cleavage products. Instead, the radiation-crosslinkable, in particular acrylic, prepolymers undergo almost quantitative crosslinking with one another and, where appropriate, also with reactive solvents present.

By using radiation-crosslinkable, in particular acrylic, prepolymers, it is possible to produce heat-resistant cling-fastener parts which can even be used as cling-fastener parts for grinding wheels or other tools, for example. Particularly high heat resistance is possessed by the highly crosslinked acrylic polymers prepared by radiation curing of formulations which encompass bi- and/or trifunctional prepolymers and/or monomers, where these promote formation of crosslinking sites. These polymers, which unlike the known

polyolefins, polyamides and polyesters can be used even at temperatures above 300°C, are essentially thermosets.

5 It is also possible to prepare polymers with predominantly thermoplastic properties via suitable selection of each of the radiation-crosslinkable prepolymers, and, where appropriate, monomers, by increasing the proportion of monofunctional prepolymers
10 and, where appropriate, monomers.

The properties of the polymers are, of course, also dependent on the chain length and the degree of crosslinking of the prepolymers used.

15 Examples of radiation-crosslinkable, in particular acrylic, prepolymers which may be used are polyester acrylates, epoxy acrylates, polyether acrylates, silicone acrylates, and urethane acrylates.

20 The use of urethane acrylates is preferred, since these are radiation-crosslinkable without inert atmosphere. Preferred urethane acrylates are the aliphatic mono-, bi- or trifunctional urethane acrylates, the aliphatic groups contributing to the flexibility of the plastic.
25 It is preferable to use bifunctional aliphatic urethane acrylates. In principle it is also possible to make at least some use of aromatic urethane acrylates of varied functionality. The viscosity of the prepolymers used
30 should preferably be from 3,000 to 60,000 mPa.s.

35 Other radiation-crosslinkable prepolymers may moreover be used in the formulation. Use of an inert atmosphere and/or an inert gas also permits the use of the following prepolymers:

1. polyester resins or chlorinated polyester resins,
or
2. utilizing a cationic crosslinking mechanism

- a) cycloaliphatic epoxy resins, or
- b) epoxy/polyol blends.

When using radiation-crosslinkable, in particular
5 acrylic, prepolymers the relatively high viscosity
mostly requires dilution of the formulation by adding
reactive diluents, in particular monomers, to achieve a
suitable viscosity. The hardness, degree of
10 crosslinking, and flexibility of the polymeric final
product, and also the viscosity of the starting
formulation, may be adjusted via suitable selection of
the monomers added.

During the polymerization, the monomers are
15 incorporated into the network, and there is therefore
almost no release of solvents from the polymer.

When using acrylic prepolymers, the monomeric reactive
diluents used are preferably acrylates of varied
20 functionality.

Addition of monofunctional acrylates reduces hardness,
increases flexibility, and gives the polymer good
adhesion properties. Monofunctional monomers also give
25 lower shrinkage during polymerization. In principle,
use may be made of any of the known monofunctional
acrylates. The monofunctional acrylates are preferably
selected from the group consisting of butyl acrylate,
2-ethylhexyl acrylate, hydroxyethyl acrylate,
30 hydroxypropyl acrylate, 4-hydroxybutyl acrylate, ethyl
diglycol acrylate, isodecyl acrylate and 2-ethoxyethyl
acrylate, particular preference being given to
ethoxyethyl acrylate and isodecyl acrylate.

35 Adding bi- or trifunctional acrylates also adjusts the
properties desired, such as hardness and flexibility.
Preferred bifunctional monomers used are diethylene
glycol diacrylate, dipropylene glycol diacrylate,
triethylene glycol diacrylate, tripropylene glycol

diacrylate, or 1,6-hexanediol diacrylate, 1,6-hexanediol diacrylate being particularly preferred.

It is also possible, if desired, to use trifunctional
5 acrylates, such as trimethylolpropane triacrylate or
pentaerythritol triacrylate, or even acrylates of
higher functionality.

It is also possible to use propoxylated monomers, which
10 are less skin-irritant.

It is preferable to use a monomer mixture made from
mono- and bifunctional acrylates, in particular a
mixture made from 2-ethoxyethyl acrylate and 1,6-
15 hexanediol diacrylate. The concentration of each of the
monomers added to the formulation depends on the
formulation viscosity required, and on the desired
hardness, flexibility and adhesion properties of the
polymer, and on the reaction rate, etc.

20 Another mixture which has proven successful is that
made from monomer- and bifunctional acrylates, in
particular ethoxyethyl acrylate or isodecyl acrylate,
with trimethylolpropane triacrylate.

25 Another advantage of producing the cling-fastener parts
from radiation-crosslinkable prepolymers and monomers
is that the adhesion properties of the plastic can be
controlled via the selection of the monomers used, and
30 that it is possible to achieve sufficient adhesion of
the plastic to a desired backing without the additional
steps of surface-treatment, by corona discharge, gas
flame, or fluorination, required in the case of known
thermoplastics. This means that the process of the
35 invention saves one operation.

To achieve sufficient polymerization using UV-curable
formulations addition of a photoinitiator is required

to form the primary free radicals which start the chain reaction on excitation by UV radiation.

5 In principle, the photoinitiators used may be any of the known molecules which liberate free radicals on absorbing UV, for example an α -hydroxyketone, α -aminoketones, dimethyl ketals of benzil, bisbenzoylphenylphosphine oxides, metallocenes and derivatives of these.

10 It is particularly preferable to use a photoinitiator comprising 2-hydroxy-2-methyl-1-phenylpropan-1-one, for example Darocur 1173 from Ciba Geigy.

15 Other conventional additives, such as dyes, stabilizers, oxygen scavengers, ferrite powder, may, of course, be added to the formulation.

20 The viscosity of the radiation-crosslinkable formulation depends on the specific conditions of production, for example the nip pressure between the shaping rolls. The viscosity of the formulation should preferably be from 150 to 20,000 mPa.s, and particular preference is given to viscosities from 300 to 25 5,000 mPa.s.

30 The percentage of prepolymers added to the radiation-curable formulation depends on the viscosity required from the formulation, on the properties of the prepolymers and monomers, and on the properties desired in the plastics material to be produced. The proportion of prepolymers in the formulation is generally from about 60 to 95%, preferably about 80%.

35 There are varied uses for the cling-fastener parts produced according to the invention: the babies' diaper sector or incontinence diapers, heat-resistant cling-fastener parts for securing grinding wheels or of other tools, for securing large areas of carpet, wall

hangings, for seat coverings or seating units, packaging, or fly-exclusion mesh, or else for self-cleaning surfaces.

5 The thickness of the cling-fastener base and the number of interlocking means per cm^2 depend on the use of the finished cling-fastener parts.

Besides cling-fastener parts, the process of the
10 invention can also produce other films which encompass protruding elements or ribs on at least one side, for example riblet films. One side of riblet films has a large number of protruding elements of a type which reduces wind shear loading and/or controls the
15 separation of boundary layers. Depending on the effects desired from the surface structure, the protruding elements may be shaped like shark skin or like a lotus flower, giving a reduction in drag and/or a self-cleaning effect. Surface structures of this type are
20 described by way of example in "Biological Surfaces and their Technological Application - Laboratory and Flight Experiments on Drag Reduction and Separation Control" by D. W. Bechert, M. Bruse, W. Hage and R. Meyer in
"Fluid Mech. (1997) Vol. 338, pp. 59-87 Cambridge
25 University Press".

Riblet films of this type are likewise produced using the formulations encompassing radiation-crosslinkable, in particular acrylic, prepolymers, these being
30 similarly molded between a shaping roll and a backing roll as appropriate, and then radiation-cured, the shaping roll having a large number of cutouts complementary to the riblet structure. The riblet films which can be produced from radiation-curable
35 formulations likewise have a high production rate and exceptionally high heat resistance. Examples of uses of the riblet films are for lowering drag on aircraft or railroads, or in pipelines, for preventing icing of aircraft, or as a self-cleaning film.

The invention will now be described using examples.

Radiation-curable formulations for producing cling-fastener parts

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A. UV-curable formulations

1. 77.7% by weight of Ebecryl 4835⁽¹⁾ from UCB Chemicals, Drogenbos, Belgium
- 10 9.7% by weight of IRR 184⁽²⁾ (ethoxyethyl acrylate) from UCB Chemicals
- 15 9.7% by weight of HDDA⁽³⁾ (hexanediol diacrylate) from UCB Chemicals
- 20 2.9% by weight of Darocur 1173⁽⁴⁾ (photoinitiator, 2-hydroxy-2-methyl-1-phenylpropan-1-one) from Ciba Geigy.

The viscosity of this formulation is about 300 mPa.s.

- 20 2. 77.7% by weight of Ebecryl 4835⁽¹⁾ from UCB Chemicals
- 25 9.7% by weight of IRR 184⁽²⁾ from UCB Chemicals
- 30 9.7% by weight of TMPTA⁽⁵⁾ (trimethylolpropane triacrylate) from UCB Chemicals
- 25 2.9% by weight of Darocur 1173⁽⁴⁾ from Ciba Geigy, as photoinitiator
3. 9.7% by weight of isodecyl acrylate from UCB Chemicals may also be used in mixing specifications 1 and 2, instead of 9.7% by weight of IRR 184 from UCB Chemicals.

B. Electron-beam-curable formulation

1. 80% by weight of Ebecryl 4835 from UCB Chemicals
- 35 10% by weight of IRR 184 from UCB Chemicals
- 10% by weight of HDDA (hexanediol diacrylate) from UCB Chemicals

2. 10% by weight of isodecyl acrylate are used instead of 10% by weight of IRR 184 from UCB Chemicals, and/or 10% by weight of TMPTA⁽⁵⁾ are used instead of 10% by weight of HDDA.

5.

10 (1) Ebecryl 4835 is stated by the manufacturer to be a mixture of aliphatic urethane diacrylates diluted with 10% of tetraethylene glycol acrylate. The viscosity at 25°C is about 4,500 mPa.s. The molar mass is about 1,600 g/mol.

15 (2) IRR 184 is a 2-(2-ethoxyethoxy)ethyl acrylate. The viscosity is stated by the manufacturer to be from about 2.5 to 9 mPa.s at 25°C.

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20 (3) The viscosity of the HDDA is stated by the manufacturer to be 10 mPa.s.

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25 (4) Darocur 1173 is stated by the manufacturer to have overlapping absorption bands in the region from 240 to 400 nm.

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(5) The viscosity of the trimethylolpropane triacrylate is stated by the manufacturer to be 115 mPa.s.

Two different apparatuses for producing cling-fastener parts are described below.

30 Figure 1 shows an apparatus for producing cling-fastener parts on a backing material by UV curing

35 Figure 2 shows an apparatus for producing cling-fastener parts with no added backing material by UV curing

Figure 3 shows a side view of a detail of a cling-fastener part 26 on a backing 13.

In the apparatus shown in Figure 1, the formulation 14 encompassing radiation-crosslinkable, in particular acrylic, prepolymers to be polymerized, as in mixing specification A1, A2 or A3 is applied in the form of a 5 film 15 of constant thickness d of from 12 to 50 μm , preferably 22+/- 5 μm , to a backing material 13, for example applied by a doctor 10 or by a die.

10 The backing material 13 used may be a plastics film, e.g. made from Hostaphan, a nonwoven, a textile, or any 15 other suitable backing material.

The film 15 made from the formulation to be polymerized 20 on the backing material 13 is then fed to a gap 16 between a shaping roll 11 and a backing roll 12. The 25 shaping roll 11 has a large number of radial cutouts 17, which are open at both ends. The viscous formulation is compressed through the gap 16 into the shape of a cling-fastener base 21, and in the cutouts, 30 into the shape of the interlocking means 24 (see also Figure 3) encompassing the interlock prominances 22 and interlock tips 23, and is then irradiated by UV light 19 of suitable wavelength. The interlocking means may have various shapes, for example a cross section which 35 is round, triangular, rectangular, pentagonal or hexagonal. The interlock tips 23 may also have a variety of shapes, and may be plate-shaped, mushroom-shaped, arched or hook-shaped, for example. Corresponding embodiments are described in DE 198 28 856.5, which is a subsequent publication.

Absorption of the UV light causes the UV-sensitive photoinitiator to liberate free radicals which initiate the free-radical chain polymerization.

35 The rate of the polymerization reaction is exceptionally high, and in a fraction of the conventional full curing time the cling-fastener parts 26 composed of the cling-fastener base 21 and of the

interlocking means 24, on the backing material 13, can therefore be released from the shaping roll 11 by means of the take-off roll 20. From about 20 to 30 m of cling-fastener sheeting can be produced per minute.

5 Since the nip pressures required are lower than in the known processes, the apparatus can have wider rolls 11, 12, 20 without any change in precision.

10 The nip pressure between the rolls 11 and 12 and the UV irradiation also achieves a firm bond between the backing material 13 and the cling-fastener base 21.

15 In order that the cutouts 17 provided in the shaping roll 11 are completely filled, a slight excess of the formulation is added, so that the bank 25 of radiation-curable composition 14 always has enough starting material available to supply the cutouts 17 in the shaping roll 11.

20 The direction of turn of the backing roll 12 and of the roll 20 is opposite to that of the shaping roll 11.

25 The UV source 19 used may be a medium-pressure mercury source. However, it is also possible to use other sources of UV radiation. Depending on the band in which the photoinitiator absorbs with free-radical formation, the wavelength range used for irradiation by UV light is from 180 to 400 nm, corresponding to from about 3 to 6 eV.

30 The irradiation wavelength depends on the emission spectrum of the source of UV radiation used, and on the band in which the photoinitiator absorbs.

35 The backing material 13 used should, of course, be substantially resistant to the UV radiation. In addition, although the backing material filters and scatters the UV radiation, it has to be ensured that, within the layer to be cured, there is sufficient

formation of the primary, photochemically generated, free radicals which initiate the chain reaction.

The apparatus shown in **Figure 2**, unlike the apparatus 5 detailed in Fig. 1, serves for the production of cling-fastener parts without any added backing material. The radiation-crosslinkable formulation 14 is present in a storage container 31, and is fed via a die 32 to the gap 16 between the shaping roll 11 and the backing roll 10 12, there being a small excess of feed, as in the apparatus described in Figure 1.

Due to the high viscosity of the radiation-crosslinkable formulation, the shaping of the viscous 15 composition brought about by the pressure applied is retained until the irradiation has caused substantially complete curing and the cling-fastener parts are released from the shaping roll 11 by means of the take-off roll 20.

20 If acrylic urethanes are used as prepolymers there is no need to work in an inert atmosphere. If other radiation-curable prepolymers are used, the reaction should be carried out in an inert atmosphere, in order 25 to prevent premature chain degradation brought about essentially by oxygen.

In an apparatus for producing cling-fastener parts by electron-beam curing, use is made of an electron beam 30 source instead of the UV source 19 in the apparatuses described in Figures 1 and 2, and one of the formulations B1 or B2, for example, is used as radiation-crosslinkable composition.

35 The energy range of the electron beam is usually from 150 to 300 keV.

The production of the cling-fastener parts from radiation-crosslinkable, in particular acrylic,

prepolymers may also take place in other apparatuses operating continuously or batchwise and encompassing means of shaping, casting, and/or compression molding formulations encompassing radiation-crosslinkable, in

5 particular acrylic, prepolymers into the shape of a
cling-fastener base with interlocking means arranged
thereupon, and which encompass a source of UV radiation
or an electron-beam source for radiation curing.

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